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NUCLEAR ENERGY RESEARCH INITIATIVE PROGRAM (NERI) QUARTERLY PROGRESS REPORT - New Design Equations for Swelling and Irradiation Creep *in Generation IV Reactors*

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QUARTERLY PROGRESS REPORT

New Design Equations for Swelling and Irradiation Creep in Generation IV Reactors

Project Number: 01-137

Quarter (4) Report

January –March 2003

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NERI Quarterly Progress Report
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A. Task A: Void Nucleation(LLNL)

1.Task Status and Significant Results:

a) We have drafted the paper describing our recently-developed computational algorithm for void nucleation and growth. This will be submitted to The Journal of Nuclear Materials in the following quarter, after institutional review and release.

b) We completed calculations for two papers on our simulations of void nucleation and growth in conjunction with dislocation evolution. One paper addresses swelling behavior for models with a fixed dislocation density, and the other addresses swelling with co-evolving dislocation content. Dislocation co-evolution appears to enhance the steady swelling behavior in solution-annealed materials. Our contrasting results should help us understand how this happens. Based on preliminary analysis, it appears to work by shaping the balance of sink strengths between the stable and unstable voids and the dislocations.

c) We have refined our algorithms for introducing discrete Monte Carlo elements into our void calculations. Our early methods for controlling this process are efficient and robust for low temperatures and high dose rates. However, our original algorithm is inefficient for low dose rates and high temperatures, where essentially unlimited numbers of Monte Carlo particles are created. (This reflects that the void nucleation stage is prolonged over 10-100 dpa for these parameters.) Our new algorithm dynamically regulates the production of Monte Carlo particles and keeps the computational cost at manageable levels even at the highest temperatures.

d) Our development of the void nucleation and growth code is essentially complete. Further work will center on incorporating additional microstructural components and evolutionary processes into the overarching simulations. We do anticipate applying the existing method to other systems such as cold-worked material and ferritic steel. However, these projects are deferred to later quarters.

2. Issues/Concerns:

a) None.

b) We have completely reworked and repeated the swelling calculations described here. This is because our early version of the void code had a numerical error in the random number generator. We repaired the problem by Oct.-Dec. 2002, and repeated our calculations. The change mainly affected the high temperature results, where there is little swelling because the nucleation of stable voids is so slow. However, we now have the correct results for the void size distributions at these high temperatures.

With the corrected code, we have found that the dislocation bias factors affect incubation times versus temperature and dose in an unexpected manner. A fixed dislocation bias factor that is appropriate to the system at late times accelerates void

nucleation during the initial transient. This can yield an unusual peak in the incubation delay for intermediate temperatures. In contrast, a density-dependent bias factor tends to prolong incubation, and eliminates the peak versus temperature.

We are systematically studying this result versus temperature, dose-rate, and initial dislocation content within our void + network dislocation model. Our new survey of parametric behavior requires that we use the improved void code (with the new algorithm for creating Monte Carlo particles). Older, less-efficient versions would be unable to produce the full dataset even with months of our available computer resources.

This current survey compares two idealized approximations to the true swelling behavior. The difference points up the effect of dislocation environment and character on the bias factors and swelling behavior. Questions about the underlying bias-factor model will be fully addressed once we incorporate a more complete evolution model for loops and network dislocations into the simulations.

c) None.

d) None.

B. Task B: Dislocation Loop Formation and Dislocation Network Evolution (LLNL)

1. Task Status and Significant Results:

a) It is now well recognized that interstitials left behind by the irradiation cascade lie mostly in clusters instead of as single point defects. Thus, it is crucial to employ dislocation evolution models with direct agglomeration of interstitial clusters. A representative distribution of pre-formed loop sizes will be needed to establish the parameters for interstitial and vacancy production in the rate-theory modeling. Some of these parameters are obtainable from existing molecular dynamics cascade simulations. We also have access to large-scale MD calculations that can be applied to the problem of compiling pre-formed cluster size distributions for appropriate systems.

b) If irradiation damage is not introduced as monomers, then dislocation climb due to reactions between dislocations and mobile interstitials must be partially replaced by dislocation-loop and loop-loop reactions involving mobile interstitial clusters. As in any reaction-diffusion theory, we must estimate loop mobilities (by glide and pipe diffusion), interactions, and barriers and pathways to collision and coalescence. The internal degrees of freedom of the dislocation loops and the low symmetry of most two-loop geometries complicate this estimation process. We have developed a loop-loop interaction model that allows for rotation of loop normal vectors as they come together. The capture volume of a large perfect loop for smaller ones is calculated by integrating the capture distances in different directions. We have found that the loop capture distance depends on torques deriving from the stress field. The net torque is altered by external stresses; this coupling to applied stress favors dislocation sinks of particular burgers vectors; and it can thereby contribute to irradiation creep.

c) We require an efficient computational method for evolution of the loop size distribution under irradiation. Loop growth from a supersaturated atmosphere of monomers is easily accessible to our existing cluster nucleation and growth simulations. However, the existing algorithm is not applicable when damage is introduced as pre-formed clusters, or when clusters are allowed to directly fuse with one another.

During this quarter, we have written a computer code to include the effects of defect-cluster coalescence on the cluster size distribution. Our rate-theory treatment makes use of the VODE package. We have applied the method to preliminary calculations of a model for irreversible cavity growth (helium bubbles). Our results show the practical importance of coalescence in creating the very largest clusters, causing a substantially broader size distribution. Coalescence also reduces the number density of large clusters, so that the overall sink strength is reduced. This alters the predicted monomer concentration and prolongs the period of efficient nucleation. We have not yet analyzed these results in detailed comparison to the Schaldach/Wolfer calculations reported in our annual report. Instead, we are focused on developing improved methods for faster calculations.

2. Issues/Concerns:

a) Extensive MD simulations of cascade debris and cascade-cluster interactions are not encompassed by this proposal. However, the ability to perform these calculations is present (at LLNL), and these questions about cascade damage are topical for other, ongoing projects. We envision obtaining enough data about irradiation cascade behavior to support our NERI modeling efforts in irradiation swelling.

b) None.

c) The VODE rate-theory method is too slow to stand alone in a full computational treatment. This is expected from our experience with the void nucleation and growth problem in the absence of cluster-cluster coalescence. There, we also found it necessary to include a Monte Carlo treatment of large clusters. A companion Markov Monte Carlo procedure for coalescence has already been identified and implementation is underway. Algorithmic development will be completed in the next quarter. The new method will be incorporated in the void nucleation and growth code, where it will be used to model void, bubble, and loop growth and coalescence. Furthermore, a parallel version of VODE exists, so we expect that our existing code can be easily extended to model cluster coalescence, if the hybrid approach proves to be impractical.

C. Task C: Dislocation-Void Interaction (UC-Berkeley and LLNL)

1. Task Status and Significant Results:

We have continued our development of a level set method approach to modeling the climb of dislocations between voids. To date, we have successfully modeled the curvature driven motion of the dislocations using a lower-order method. The true dynamics of the dislocations, however, require motion of the dislocation based on the Laplacian of the curvature (a first step towards moving the dislocations in response to the Laplacian of the chemical potential). The lower-order method proved incapabable of modeling this more complicated situation. We are presently developing a higher order level set method in order to allow modeling of the proper dislocation dynamics.

2. Issues/Concerns:

None.

D. Task D: Model Integration (LLNL)

1. Task Status and Significant Results:

a) The newly installed VODE package functionally replaces the old Euler-based rate-theory solver in our void nucleation and growth code. It is slower than the older scheme. However, the new method makes it possible to perform nucleation and growth simulations without the quasi-stationary approximation for monomers. We deem this improvement to be of low priority, and so we defer a systematic study of its effect until a later date. However, this improvement also allows the generation of irradiation damage as pre-formed clusters rather than simply as free monomers. We are now able to incorporate any desired distribution of pre-formed cluster sizes. This is considered a significant development, as it will refine our study of loop evolution and its effect on swelling behavior. For example, it is now easy to introduce vacancy defects as immobile, pre-formed clusters. They can evaporate at moderate temperatures to create the vacancy supersaturation that drives void growth. The added evaporation step will alter temperature-dependences of incubation and swelling alike.

b) When finished, the coalescence simulation capability described in Task B will be important to incorporating loop and network dislocation evolution into our overarching microstructure simulations. Loops containing several thousand vacancies or interstitials have sizes approaching the average separation of network dislocations. In that limit, they can be treated as part of the network, independent of their (ideally-circular) radius. This maximum size limit greatly simplifies the computational problem, although it is still out of reach of a pure, single-processor VODE calculation.

2. Issues/Concerns:

a) Evaluation of production bias effects and pre-formed cluster generation should be informed by realistic distributions of cluster sizes. We assign this problem to Task B. However, studies can also proceed with some idealized size distribution in order to reveal qualitative changes in the predicted temperature- or flux-dependence of the swelling. Testing of pre-formed clusters in our model and in our computer code is not contingent upon a complete MD survey of cascade cluster distributions.

b) Realistic simulation of pre-formed loop evolution requires a complete model treatment of loop-monomer interaction and loop mobility and loop-loop coalescence. We are developing such a model in Task B. However, once again, simplified, "toy" models may be sufficient to reveal the direction of changes to the swelling behavior. We expect to be able to evaluate loop coalescence effects before the final loop-loop model is established.

Currently, additional algorithm development is underway for a hybrid rate-theory/Monte Carlo treatment of the coalescence simulations. Based on the void growth results in hand, we expect the hybrid method to be practical on single-processor computers.

E. Task E: Validation (PNNL and LLNL)

1. Task Status:

a) We have used the full set of microstructural data from a pure ternary alloy

irradiated at different neutron fluxes to test the fundamental theory of irradiation swelling. The standard rate theory links the swelling rate to the microstructure and the materials properties. The simplest version attempts to predict the swelling rate using essentially two equations. We have found that swelling calculations based on standard rate theory with reasonable parameter values agree quite well with the experimental results. These results further validate the use of models based on the classical rate theory of irradiation swelling. This critical comparison does not attempt to predict the microstructural evolution; it only shows that if the microstructure is known then the instantaneous rate of swelling can be explained by the classical theory. In addition, we have narrowed the dislocation bias factor ratios from a maximum possible range of 1.1 - 2.0 to 1.25-1.55. This is consistent with microscopic materials theory estimates.

b) We have largely completed the analysis of two identical sets of special austenitic steels irradiated in the Russian reactors BOR-60 and BR-350. The samples encompass 4 levels of cold work, 3 different solute additions (Nb, Ti, and Nb-Ti) and exposures at high and low flux at the similar temperatures (470-480 C vs. 490 C) and to the same total fluence (53 dpa). The low-flux exposures reach greater swelling, by factors of 2.5 to 8.5. Swelling is always greater at the lower flux, even though the interaction of flux and material dependences can be complex. These results broaden and reinforce our focus on incubation dose-rate dependence.

c) We are analyzing the "scatter" in existing swelling data to determine if the deviations are due to previously unrecognized parametric dependences. The incubation dose-rate effect that we are studying confounded early data analysis until it was realized that incubation and steady behaviors were being confused. Similarly, the unexpected peak in simulated incubation times at intermediate temperatures would likely appear as "scatter" in the existing experimental databases.

2. Issues/Concerns

a) None.

b) None.

c) Statistical analysis of experimental datasets are complicated by the strong correlations between, e.g., temperature and flux within a given experiment.

F. Task F: Development of Constitutive Equations (LLNL and PNNL)

1. Task Status:

We have developed a simplified three-component model of irradiation swelling, based on our numerical simulations. The classical picture holds that the swelling rate reaches a maximum value when the sink strengths of dislocations and voids are comparable. Simulations of the full void size distribution do not reproduce this relation. Instead, the total void sink strength often reaches much larger values than the dislocations, before the peak rate of swelling is achieved. We find that the discrepancy is due to the presence of numerous, small, unstable vacancy clusters that are formed and are sustained by the vacancy supersaturation. Once the nucleation of stable voids is complete, these unstable clusters essentially neither grow nor shrink on average – they reach a quasistationary distribution. Therefore, these clusters absorb equal numbers of vacancies and interstitials over time, and so they act as indirect recombination sites. The flux of defects to the three sinks of dislocations, stable voids, and unstable clusters

completely dictates the temperature-, flux-, and dose-dependent rate of swelling.

Replacing the many thousands of different-sized void sinks with just two parameters supports the use of simplified microstructures for constitutive models of irradiated materials. Our results appear to be consistent with the classical theory analysis of experimental microstructures in Task E of this report. The materials parameters chosen for that critical analysis included a vacancy-interstitial recombination radius. It is found that this parameter may have larger values than are predicted from a microscopic theory of vacancy-interstitial annihilation. This would be consistent with an additional, indirect recombination process at the unstable clusters.

2. Issues/Concerns:

None.

Status Summary of NERI Tasks – Phase 1-3:

Phase1:

Milestone/Task Description	Planned Completion Date	Actual Completion Date
Void nucleation with disloc.	Sept. 2002	June 2002
Disl. Loop formation	Sept. 2002	Sept. 2002

Phase2:

Milestone/Task Description	Planned Completion Date	Actual Completion Date
Preformed defect-clusters & Void Formation	August 2003	postponed to Phase 3.
Incubation for SS304 & Pure Alloys	May 2003	Mar. 2003
Coupled Loop and Network Evolution	June 2003	postponed to Phase 3
Cluster coalescence modeling (He bubbles)	Sept. 2003	
Tabulation of MD cascade debris cluster distributions	Sept. 2003	
Functional Forms for Constitutive Equations	Sept. 2003	

Phase 3:

Milestone/Task Description	Planned Completion Date	Actual Completion Date
Joint bubble-void simulations	Dec. 2003	
Void growth with preformed loop defects	Dec. 2003	
Void growth plus loop growth simulations	Feb. 2004	
Joint bubble/void loop/dislocation simulations	May. 2004	